Manipulation of the Spontaneous Emission Dynamics of Quantum Dots in 2D Photonic Crystals

A. Kress,* F. Hofbauer, N. Reinelt, M. Kaniber, H. J. Krenner, R. Meyer, G. Böhm, and J. J. Finley

Walter Schottky Institut and Physik Department, Technische Universität München, Am Coulombwall 3, D-85748 Garching, Germany

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Abstract

We demonstrate the ability to control the spontaneous emission dynamics of self-assembled quantum dots via the local density of optical modes in 2D-photonic crystals. We show that an incomplete 2D photonic bandgap is sufficient to significantly lengthen the spontaneous emission lifetime ($> 2\times$) over a wide bandwidth ($\Delta\lambda \geq 40$ nm). For dots that are both *spectrally* and *spatially* coupled to strongly localized ($V_{mode} \sim 1.5(\lambda/n)^3$), high $Q \sim 2700$ optical modes, we have directly measured a strong Purcell enhanced shortening of the emission lifetime $\geq 5.6\times$, limited only by our temporal resolution. Analysis of the spectral dependence of the recombination dynamics shows a maximum lifetime shortening of 19 ± 4. From the directly measured enhancement and suppression we show that the single mode coupling efficiency for quantum dots in such structures is at least $\beta = 92\%$ and is estimated to be as large as $\sim 97\%$.

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*Electronic address: kress@wsi.tum.de

Designer photonic materials fabricated from periodic dielectrics provide a direct route toward achieving complete control of the spontaneous emission of solids.[1, 2] The ability to manipulate the strength of the light-matter coupling in this way lies at the very heart of modern optics, with a variety of potential applications ranging from integrated photonics[3] to fundamental quantum optics.[4, 5, 6, 7] One of the most remarkable quantum optical phenomena is the deterministic generation of single photons from isolated quantum emitters. In this context, individual semiconductor quantum dots (QDs) are ideal solid state emitters due to their high radiative efficiency, stability and ease of incorporation into active devices.[8, 9, 10, 11] However, for QDs embedded within an isotropic semiconductor the single photon extraction efficiency is extremely low ($\eta_{ex} < 1\%$) limiting their realistic potential for applications in quantum information science.[5, 12] This problem can be addressed by locating QDs within optical cavities and utilizing the Purcell effect[13] to funnel single photons into a single optical mode for collection.[10] Using such approaches, η_{ex} larger than a few percent have been reported for single dots incorporated into pillar microcavities.[10] Cavities realized using photonic crystals (PCs) may provide maximum flexibility to tune the local density of photon states over a much wider bandwidth and achieve full control of the spontaneous emission via the strength of the local vacuum field fluctuations.[2] Furthermore, strongly localized modes in PCs combine a planar geometry with high quality factors ($Q = \omega\tau_{photon}$) and small effective mode volume (V_{eff})[15, 16, 17], potentially advantageous properties for achieving strong Purcell enhancement and realizing *efficient* QD based single photon emitters.

In this paper we demonstrate control of the QD spontaneous emission dynamics in such 2D PCs. For dots that are both *spectrally* and *spatially* coupled to high Q, strongly localized cavity modes we directly measure a pronounced shortening of the emission lifetime ($\geq 5.6\times$), limited only by the temporal resolution of our detection system. Analysis of the spectral dependence of the decay rate as a function of emitter-cavity detuning shows that the maximum enhancement is as large as 19 ± 4. A strong ($> 2\times$) reduction of the emission decay time is observed over a wide bandwidth for dots detuned from the cavity modes, demonstrating that a partial, TE-bandgap is sufficient to tailor the QD spontaneous emission dynamics. From the directly measured enhancement and suppression of the spontaneous emission lifetime we extract a single mode coupling efficiency for dots in these PCs of $\beta = 92\%$, the spectrally dependent measurements indicating that it may become as large as $\beta \sim 97\%$.

The samples investigated consisted of a $d = 400$ nm thick Air-GaAs-Air slab waveguide into which a 2D photonic crystal (PC) is defined by fabricating a triangular lattice of air holes in the GaAs waveguide and defining a suspended membrane by removing an underlying AlAs layer using wet chemical etching. A single layer of nominally $In_{0.5}Ga_{0.5}As$ QDs was incorporated into the center of the GaAs waveguide core as an internal light source. Low mode volume microcavities were formed by introducing single missing hole point defects in the hexagonal lattice of holes, realizing $H1$ resonators. The PCs have a periodicity of $a = 300$nm and the air hole radius (r) was varied to tune the cavity mode energies through the inhomogeneously broadened spectrum of QD groundstates and control the width of the photonic bandgap. From a QD areal density of $\sim 200\mu m^{-2}$ and the detection spot size of $\sim 1\mu m^2$ we estimate that only a small number of dots (~ 5) are spectrally coupled to the high Q cavity modes.

Spatially resolved optical measurements were performed at $T = 10K$ using a confocal micro photoluminescence (μPL) system that provides a spatial resolution $\sim 1\mu m$. For CW measurements, the samples were excited using a HeNe laser and the resulting PL signal was dispersed by a 0.55m imaging monochromator and detected using a nitrogen cooled Si-CCD camera. Time resolved measurements were performed by exciting the sample using $\sim$ 50 ps duration pulses at $\lambda = 658$ nm and detecting the temporal decay of the resulting luminescence using a single photon Si-avalanche photodetector and time correlated photon counting electronics. The maximum temporal resolution provided by this system is $\sim 150ps$ after deconvolution with the system response function, much shorter than the intrinsic ground state radiative lifetime of our QDs ($\tau_0 \sim 0.8ns$).

Figure 1a compares an ensemble PL spectrum with μPL spectra recorded from a series of $H1$ cavities as r/a is increased systematically from 0.33−0.42. Over this range of parameters, the μPL spectra reveal a prominent doublet, labeled $M1$ and $M2$ in Fig. 1, corresponding to dipole like cavity modes orientated along the $\Gamma - M$ and $\Gamma - K$ crystal directions (see inset - Fig. 1c). Under the present strong excitation conditions ($P_{ex} \sim 100Wcm^{-2}$) the PL intensity is determined by the QD spontaneous emission lifetime and a 50× enhancement of the PL intensity is observed for dots spectrally on resonance with the cavity modes when compared with dots that are detuned. This observation indicates the presence of pronounced cavity QED effects, an expectation confirmed by our time resolved measurements presented below.

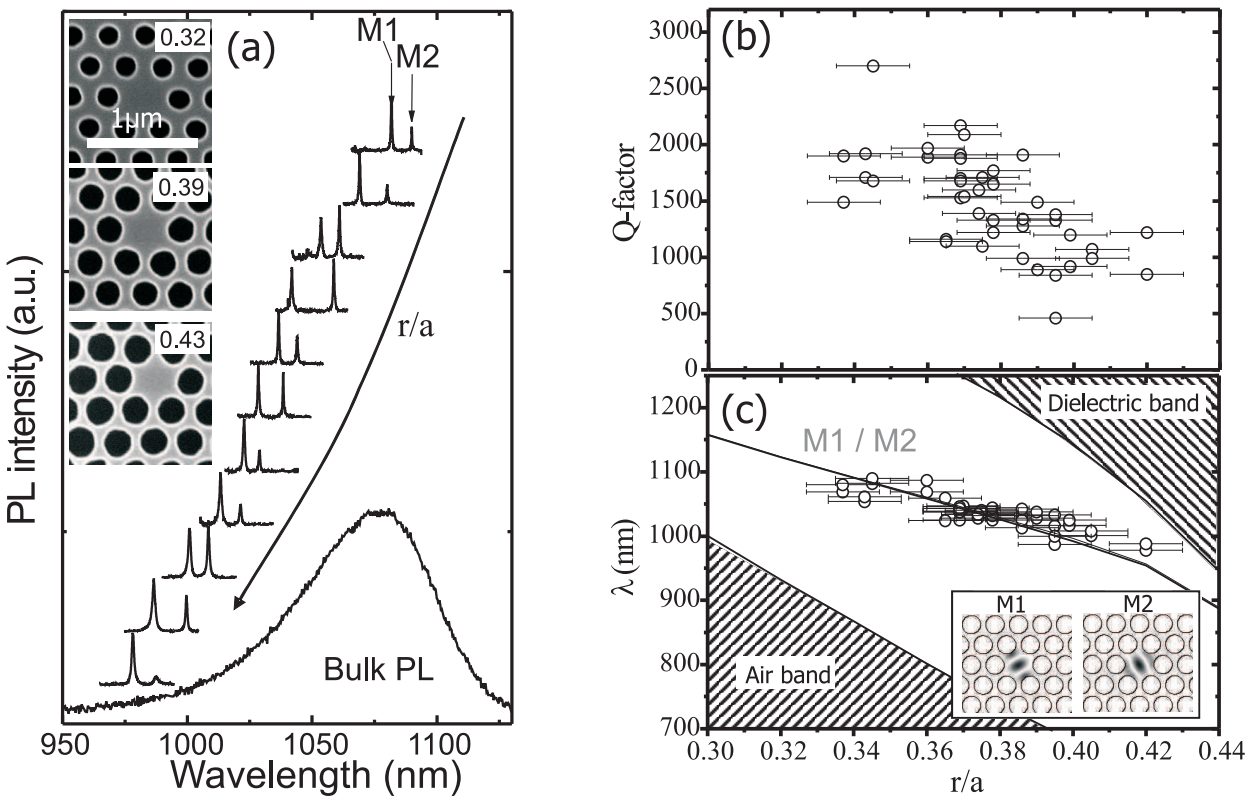

FIG. 1: (a) PL spectra recorded from a series of $H1$ PC cavities as a function of the ratio of hole radius r to periodicity a. (inset) The SEM images show typical cavities. The ensemble PL is shown for comparison. (b) Measured Q factors for the cavity modes of > 40 cavities as a function of r/a. (c) Calculations of the $TE-$polarized $3D$ bandstructure with $a = 300$ nm showing the localized dipole like modes $M1$ and $M2$ inside the photonic bandgap and the experimental data. (inset) Calculated electric field profile of the cavity modes.

Fig. 1b shows the cavity mode Q-factors deduced for over 40 structures plotted as a function of r/a. A significant increase of Q from ~ 500 to ~ 2700 is observed as r/a is reduced from 0.42 to 0.33. This can be explained by considering the position of the cavity modes within the TE-photonic bandgap.[16] Fig. 1c shows the calculated bandstructure for our structures as a function of r/a.[18] The continuum *dielectric* and *air* bands are marked by the shaded regions, together with the TE photonic bandgap and the $M1$ - $M2$ doublet (solid lines).[19] The calculated wavelength of the cavity modes and its dependence on r/a are in good quantitative agreement with our measurements, confirming their identification.[20] The calculations presented in Fig. 1c show that $M1$ and $M2$ shift progressively deeper into

the photonic bandgap as r/a is reduced. As a consequence, the modes couple more weakly to the dielectric band continuum resulting in the observed enhancement of the Q-factor.[16] We now shift our attention to the emission dynamics of QDs whose emission frequency lie throughout the TE-photonic bandgap, both in and out of resonance with the highest Q cavity modes.

The maximum photon lifetime in our cavities is $\tau_{photon} = Q_{max}/\omega \sim 2$ ps, much shorter than the typical QD spontaneous emission lifetime ($\tau_0 \sim 0.8$). Furthermore, since the QD homogeneous linewidth is much narrower than the cavity mode ($\Delta\lambda_c = \lambda_c/Q \sim 0.5nm$, c.f. $\Delta\lambda_{QD} \ll 0.1nm$ [21]) the light matter coupling remains in the perturbative regime and can be described by the Purcell effect.[13, 14] In this case, for an ideal emitter on resonance with the cavity mode the spontaneous decay lifetime is reduced by a factor $F_p = 3Q/(4\pi^2 V_{mode})$, where V_{mode} is the effective volume of the cavity mode in units of $(\lambda_c/n)^3$. For the cavities discussed here, we calculate $F_p \sim 100$, for $Q \sim 2000$ and $V_{mode} \sim 1.5(\lambda/n)^3$, in good accord with the $\sim 50\times$ total enhancement of the emission intensity observed for QDs spectrally in resonance with the cavity modes (see Fig. 1a). However, to unambiguously separate the influence on the QD radiative lifetime of the local density of photonic states from simple improvements of the collection efficiency due to the PC, we performed time resolved measurements.

A PL spectrum from the cavity selected for time resolved studies is presented in Fig. 2a, showing cavity modes suitable for detection using our silicon based detection system (λ_{M1} =1025.4 nm with Q_{M1} =1500 and λ_{M2} = 1031.5nm with Q_{M2} =1950). We compared μPL decay transients recorded both *in* and *out* of resonance with the cavity modes with the *intrinsic* QD dynamics measured on the unpatterned GaAs membrane without the PC (τ_0). Figure 2b compares raw time resolved data recorded from QDs in the cavity, but strongly detuned from the cavity mode (filled circles) with reference data recorded at the same wavelength (λ_{det1} = 1037 nm) from the unpatterned membrane (open circles). For both transients, we observe monoexponential decays with time constants of $\tau_1 = 1.8 \pm 0.1$ ns and $\tau_0 = 0.84 \pm 0.05$ ns, respectively. The QDs located within the cavity have much longer decay times ($\tau_1/\tau_0 \sim 2$) when compared with dots in the pure membrane, indicating the presence of a gap in the local photonic density of states due to the 2D photonic bandgap.[2] This suggestion is further substantiated by our spectrally resolved measurements presented below.

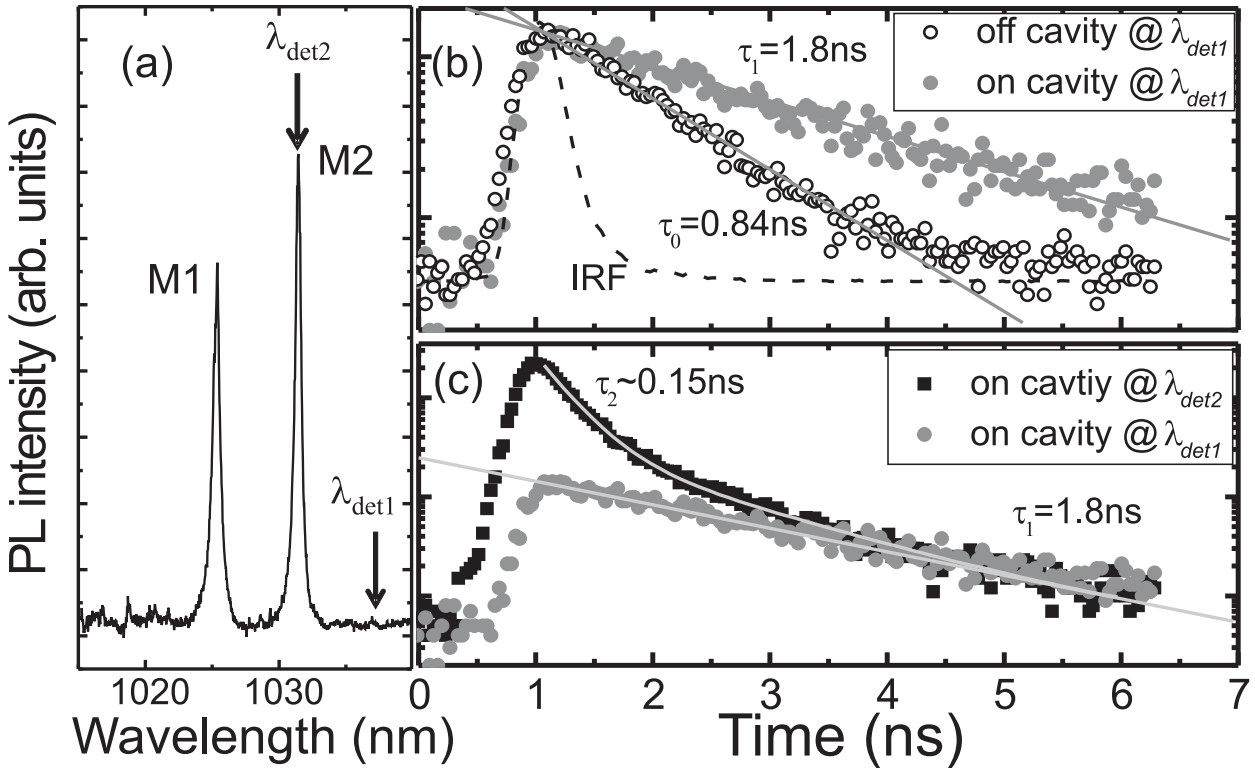

FIG. 2: (a) PL spectrum of the selected cavity. (b) Comparison of decay transients recorded from QDs detuned from the cavity modes (λ_{det1}=1037nm) either (i) within the unpatterned GaAs membrane (τ_0 - open circles) or (ii) from the PC (τ_1 - filled circles). The dashed line is the instrument response function of our setup (IRF). (c) Decay transients recorded from the PC $H1$ cavity both *in resonance* with $M2$ at λ_{det2}=1031.5 nm (filled squares) and detuned at λ_{det1}=1037nm (filled circles).

Figure 2c compares a decay transient recorded from dots in the cavity, here recorded in-resonance with $M2$ (filled squares), with the data of Fig. 2b detuned strongly by $\sim$ 5nm to longer wavelength (open circles). In contrast with all dynamics discussed until now, the decay transient recorded on resonance with $M2$ shows a clear double exponential decay, as confirmed by a fit of $I(t) = A\exp(-t/\tau_2) + B\exp(-t/\tau_1)$ shown on the figure. The longer time constant ($\tau_1 = 1.8 \pm 0.1$ ns) is identical to that discussed above for QDs spectrally detuned from the cavity mode, whereas the faster transient ($\tau_2 \sim 0.15ns$) is limited by the time resolution of our setup. We identify this behavior as arising from a strong Purcell enhanced shortening of the emission time, compared with τ_0, measured for dots that are

both *spectrally* and *spatially* on resonance with the cavity mode. From figure 2b and 2c we obtain already a factor $\tau_0/\tau_2 \geq 5.6 \pm 0.3$, while the decay time τ_2 is limited by the system time resolution (see instrument response function in Fig. 2b). The longer decay transient τ_1 is identified as arising from QDs that are spectrally on resonance with $M2$ but do not couple to the cavity mode due to their position outside the cavity in the body of the PC. From these directly measured decay times for coupled (τ_2) and uncoupled (τ_1) dots (see figure 3) we obtain ratios of $\tau_1/\tau_2 = 12 \pm 1$, defining a single mode coupling efficiency $\beta = 1 - (\tau_2/\tau_1) \sim 92\%$ for dots placed both spectrally and spatially on resonance with the cavity mode. This figure of merit provides significant promise for the realisation of efficient, QD based, single photon sources based on PC cavities.[3, 10, 11]

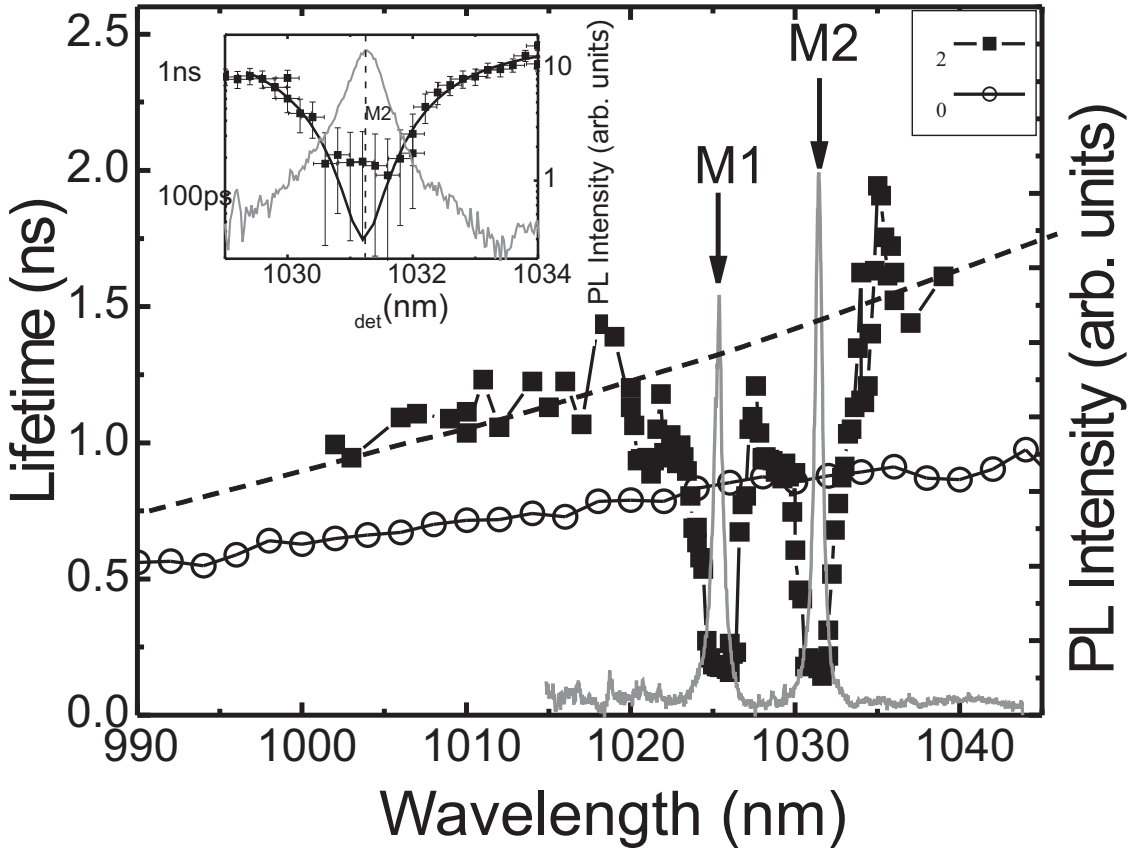

FIG. 3: Spectral dependence of the decay lifetimes for dots within the PC (filled squares) and within the unpatterned membrane (open circles). The dotted line represents a guide to the eye showing the stronger suppression of the spontaneous emission for wavelengths closer to λ_{mid}^{TE}. (inset) Fit of eqn 1 to the spectral dependence of the decay lifetime.

Figure 3 compares the spectral dependence of the QD decay time in the membrane, but away from the PC (τ_0 - open circles) and the dominant decay time for dots in the cavity (filled squares), representing the faster of the two time constants extracted from the biexponential fit. A reference PL spectrum is also presented for comparison. The intrinsic QD lifetime τ_0 does not vary between QDs in the unprocessed material and in the unpatterned GaAs membrane. It increases weakly from $\tau_0 \sim 0.65 - 0.90ns$ as the detection wavelength increases from $1000 - 1040nm$. From the data presented Fig. 3 the significant lengthening (off-resonance) and shortening (on-resonance) of the decay liftime discussed above can be clearly observed. Moreover, the lengthening is found to occur over a remarkably wide bandwidth ($\Delta\lambda \geq 40nm$) and becomes more pronounced towards longer wavelength as shown schematically by the dotted line on Fig. 3 that acts as a guide to the eye. For the presently investigated PCs, the middle of the TE-polarized photonic bandgap lies at $\lambda_{mid}^{TE} \sim 1100$ nm (see Fig. 1c) and there is no overlapping gap for both TE and TM polarized waveguide modes. The observed spectral dependence is attributed to a progressive reduction of the local photon density of states as the wavelength approaches λ_{mid}^{TE}, indicating that even a *partial* 2D photonic bandgap is sufficient to significantly inhibit spontaneous emission. We believe that this is due to the predominantly *heavy* hole character of the QD ground state exciton transition[22] that gives rise to TE polarised emission. Therefore, we suggest that tailoring of only the TE-optical modes is sufficient to strongly modify the spontaneous emission properties of self-assembled QDs in 2D-PC nanocavities.

The minima in the spectral dependence of the decay lifetime ($\tau(\lambda)$) close to $M1$ and $M2$ are $\sim 4\times$ broader than the cavity modes in the emission spectrum. Since the Lorenztian cavity modes should lead to a similar spectral profile in $\tau(\lambda)$, this observation indicates that the reduction of the decay time for zero detuning is much larger than the measured ~ 150 ps, limited by our temporal resolution. In the weak coupling regime photon reabsorption can be neglected and Fermi's golden rule provides the spontaneous decay time relative to its value in a homogeneous medium τ_0/τ_2.[23]

$$\frac{\tau_0}{\tau_2} = \frac{1}{3} F_P \frac{\left|\vec{E}(\vec{r})\right|^2}{\left|\vec{E}_{max}\right|^2} \frac{\Delta\lambda_{cav}^2}{\Delta\lambda_{cav}^2 + 4(\lambda_{cav} - \lambda_{QD})^2} + \alpha \qquad (1)$$

In equation 1 λ_{QD} and λ_{cav} are the QD and cavity wavelength and $\Delta\lambda_{cav}$ is the linewidth of the cavity mode measured from the PL spectrum. Two different decay channels are

taken into account in Eqn. 1; the first term describes the spontaneous emission of a dot located at $\vec{r}$ into the cavity mode with an local electric field $\vec{E}(\vec{r})$ and a maximum amplitude $\vec{E}_{max}$, whereas in the second term α describes a possible decay channel due to emission into residual modes in the quasi-photonic bandgap. By fitting Eqn. 1 to the measured $\tau(\lambda)$ data we extract the decay time on resonance, the best fit is compared with the PL intensity in Fig. 3(inset) on a logarithmic scale. The fitted spectral dependence of the decay time, now has exactly the same lineshape as the PL intensity but with a much shorter decay time $\tau_2 = 44 \pm 8ps$ on resonance. From the fit, we estimate a maximum shortening of the decay time by a factor $\tau_0/\tau_2 = 19 \pm 4$ for ideally located QDs on resonance, corresponding to a maximum Purcell factor $F_P = 56 \pm 10$. This value is in fairly good agreement with the maximum value of ~ 100 calculated from the measured Q and V_{eff}, the discrepancy probably arising from a displacement of the QDs probed relative to the electric field antinode in the cavity. For the obtained values of $\tau_1 = 1.8ns$ and $\tau_2 \sim 50ps$ we estimate a maximum single mode coupling efficiency for this system of $\beta \sim 97\%$.

In summary, we have investigated the influence of the modified density of states in PC microcavities on the spontaneous emission dynamics of self-assembled QDs. A reduction of the spontaneous emission lifetime up to $(5.6 \pm 0.3)\times$ was directly measured for dots on resonance with the cavity modes and a lengthening $> 2\times$ off resonance over a bandwidth ≥ 40 nm. From the spectral dependence, a maximum enhancement of τ_0/τ_2 up to 19 ± 4 was deduced, corresponding to a drastic shorting of the exciton lifetime to only $\sim$ 50ps. This indicates that the single mode coupling efficiency may become as large as $\beta \sim 97\%$. Finally, since the numbers reported here are ensemble averages, they may become even larger for an ideally placed dots with a dipole moment aligned perfectly with the cavity field. Therefore, the present results provide significant promise for realisation of *efficient* single photon emitters based on PC nano-cavities.

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